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A Kinetic Monte Carlo method for the simulation of heteroepitaxial growth

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Abstract

We introduce a simulation algorithm which allows the off-lattice simulation of various phenomena observed in heteroepitaxial growth (see e.g. [Politi et al., Phys. Rep. 324 (2000) 271–404]) like a *critical layer thickness* for the appearance of misfit dislocations, or *self-assembled* island formation in $1+1$ dimensions. The only parameters of the model are deposition flux, simulation temperature and an interaction potential between the particles of the system.

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Keywords: Kinetic Monte Carlo simulation; Heteroepitaxial growth; Misfit dislocations; Critical thickness; 2D–3D transition

1. Introduction

Common methods for the simulation of heteroepitaxial growth [1], like molecular dynamics, consume a lot of computer time and are therefore only practicable for rather small system sizes (see e.g. [2]). On the other hand a lot of parameters are needed to include stress in faster methods like Kinetic Monte Carlo simulations (KMC) with a fixed lattice.

Here, we propose a KMC algorithm for the simulation of the early stages of heteroepitaxial growth in $1+1$ dimensions. In contrast to similar off-lattice algorithms suggested before—for example, by Faux et al. [3,4], Plotz et al. [5] or Schindler [6]—we are able to simulate heteroepitaxial growth for rather thick adsorbate layers and over a wide range of the misfit between the lattice constants of substrate and adsorbate. The only parameters of the model are the deposition flux R_d, temperature T and a *Lennard–Jones* interaction potential between the particles of the system.

Using this algorithm we are able to determine the dependence of the *critical layer thickness* for the appearance of misfit dislocations on the misfit ε between substrate and adsorbate (see also [7]).

Furthermore, we find a 2D–3D transition at the formation of islands depending on the misfit ε and temperature T during the early stages of growth. This transition is identified with the self-assembled island formation in heteroepitaxial growth (see e.g. [8]). We observe that this transition takes place at a distinct 2D island size and discuss the dependence of the transition size on the misfit ε.

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2. Method

The proposed simulation algorithm for heteroepitaxial growth combines Kinetic Monte Carlo simulations (KMC) with a method for energy barrier calculation.

The two-dimensional simulation cell is open in the vertical and periodic in the lateral direction. Growth proceeds on a six atomic layers thick substrate with fixed bottom layer. The system size L is given by the number of particles in the substrate's topmost layer and is chosen between $L = 30$ and $L = 200$.

Adsorbate particles are randomly deposited on the crystal surface at a rate $R_d = LF$, where $F = 1^{-1}$ s is the deposition flux. The rate R_i for a diffusion event i is given by

$$R_i = \nu_0 \mathrm{e}^{-E_{a,i}/(k_\mathrm{B}T)}, \tag{1}$$

where $\nu_0 = 10^{12}$ s^{-1}, $E_{a,i}$, T are the attempt frequency, the activation barrier for the diffusion step i and the simulation temperature, respectively.

The total rate R of all microscopic processes therefore sums up to

$$R = R_d + \sum_i R_i. \tag{2}$$

Using a binary tree structure [9] an event i is chosen with the correct probability. Then this event is performed and the rates of all affected events are updated.

Unlike in standard Monte Carlo simulations time does not advance linearly in discrete time steps Δt. Instead, the time τ between two microscopic processes is given according to a Poisson distribution $P(\tau) = R\mathrm{e}^{-R\tau}$ by $\tau = -\ln\rho/R$, where ρ is a uniformly distributed random number between 0 and 1.

As the aim is the simulation of heteroepitaxial growth it is necessary to overcome the limitations of a fixed lattice. For this reason a pair potential U_{ij} between two particles i and j which are separated by a *continuous* distance r_{ij} is introduced.

The aim here is not to model a specific material realistically but to gain general insight into relevant mechanisms of heteroepitaxial growth. We therefore choose a simple *Lennard–Jones* potential

$$U_{ij}(\sigma) = 4U_0\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right] \tag{3}$$

as particle interaction potential. However, we focus on the observation of effects which should not crucially depend on the particular choice of the potential.

The equilibrium distance r_0 between two particles interacting via U_{ij} becomes $r_0 = \sqrt[6]{2}\sigma$ which is somewhat smaller in the bulk material. Because of the isotropy of the Lennard–Jones interaction the particles arrange in a triangular lattice. In order to save computer time the interaction potential U_{ij} is cut off at a distance $r_{ij} > 3r_0$ which is justified, because interaction strength at this distance is less than 1% of the value at the equilibrium distance.

The interaction of two substrate particles is given by $U_{ij}(\sigma_S)$. Two adsorbate particles interact via $U_{ij}(\sigma_A)$ whereas a substrate and an adsorbate particle interact via $\frac{1}{2}(U_{ij}(\sigma_S) + U_{ij}(\sigma_A))$. In the following σ_S is set $\sigma_S = 1.0$ and σ_A is chosen between 0.85 and 1.15, so one can simulate heteroepitaxial growth for a misfit

$$\varepsilon = \frac{\sigma_A - \sigma_S}{\sigma_S} \tag{4}$$

between -15% and $+15\%$.

The activation barrier $E_{a,i}$ for the diffusion step i is given by $E_{a,i} = E_{t,i} - E_{b,i}$, where $E_{t,i}$ denotes the energy of the particle at the the transition state and $E_{b,i}$ the energy at the binding state. Both are calculated for a frozen crystal using Brent's method [10]. First, the potential $U_{i,x_i}(y_i)$

$$U_{i,x_i}(y_i) = \sum_{j=1}^{n} U_{ij}, \quad j \neq i, r_{ij} < r_0 \tag{5}$$

of particle i interacting with all particles within a circular region of radius $3r_0$ is minimized varying its vertical coordinate y_i and with a fixed horizontal coordinate x_i. The minimization of $U_{i,x_i}(y_i)$ with variation of x_i then leads to the energy at the binding state $E_{b,i}$. The energy at the transition state $E_{t,i}$ is calculated by a maximization of $U_{i,x_i}(y_i)$. Here the saddle point search is replaced by a maximum search, which is possible in $1 + 1$ dimensions.

To consider the elastic deformation of the crystal after each microscopic event (diffusion or deposition) the total potential energy of the n particle system

$$E_\mathrm{tot} = \sum_{i=1}^{n}\sum_{j=i+1}^{n} U_{ij} \tag{6}$$

is minimized using a conjugate gradient method [10] with variation of the coordinates of all particles within a circle of radius $3r_0$ around the particle where the event took place. In order to avoid strain caused by this local relaxation of the crystal, after a distinct number of microscopic events—depending on the misfit ε—a minimization of E_{tot} with variation of all particle coordinates is performed.

Using the described algorithm we are able to simulate heteroepitaxial growth for system sizes up to $L = 200$ and up to 30 monolayers of deposited particles within reasonable computer time.

3. Misfit dislocations

First, heteroepitaxial growth is simulated in order to determine the critical layer thickness h_c for the appearance of dislocations as a function of the misfit ε. To this end between 5 and 10 independent simulation runs are carried out for each value of ε at a temperature $T = 0.03U_0/k_{\text{B}}$.

We find that in each simulation run several dislocations appear simultaneously. Then, after the deposition of a few monolayers of adsorbate after the first appearance of a dislocations in the crystal the number of dislocations remains constant. The thickness of the adsorbate layer at which dislocations first appear is registered as h_c.

Fig. 1 shows sections of two crystals obtained in our simulations for left panel $\varepsilon = +10\%$ and right panel $\varepsilon = +6\%$. The crystal section in the left panel contains a *perfect dislocation*, indicated by a Burgers vector which is an integer multiple of a lattice vector. The right panel shows a crystal section containing a *partial dislocation*—characterized by a Burgers vector which is a rational fraction of a lattice vector.

Fig. 2 shows the critical layer thickness h_c plotted versus the absolute value of the misfit ε. For $-0.03 < \varepsilon < 0.02$ the critical thickness is too large to be observed in our simulations.

Due to the fact that the Lennard–Jones potential is steeper in compression ($\varepsilon > 0$) than in tension ($\varepsilon < 0$), the simulation results show a dependence of h_c on the sign of the misfit.

Our simulation results agree well with a power law (solid lines in Fig. 2)

$$h_c = a^* \varepsilon^{-3/2} \tag{7}$$

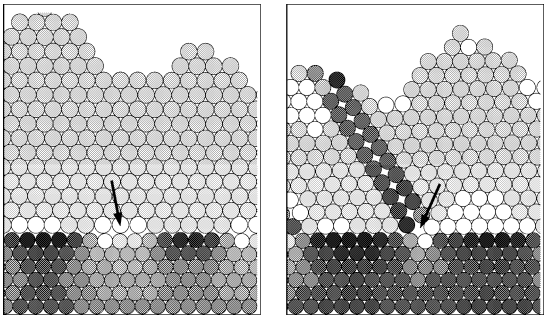

Fig. 1. Typical sections of crystals obtained in our simulations. The six bottom layers are the given substrate. The dislocations are marked with arrows. Left panel: perfect dislocation for $\varepsilon = +10\%$. Right panel: partial dislocation for $\varepsilon = +6\%$. The grey level for a particle indicates the particle's average distance to its nearest neighbors of the same kind: the lighter its grey level the more is this particle under compression.

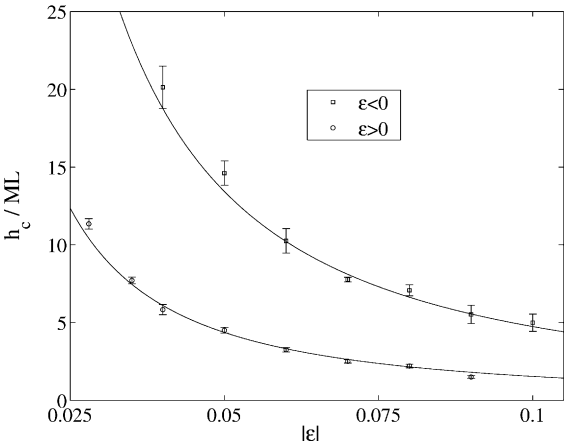

Fig. 2. Critical thickness h_c versus misfit $|\varepsilon|$ for $\varepsilon < 0$ (upper curve) and $\varepsilon > 0$ (lower curve). The error bars are obtained as the standard error of the simulation results. The solid lines are calculated using Eq. (7) where $a^* = 0.15$ for $\varepsilon < 0$ and $a^* = 0.05$ for $\varepsilon > 0$.

which was proposed by Cohen-Solal et al. [11]. A nonlinear fit of our results yields $a^* = 0.15$ for $\varepsilon < 0$ and $a^* = 0.05$ for $\varepsilon > 0$.

4. 2D–3D transition

At higher temperatures it becomes more likely for a particle to jump from or on a monolayer island because of the dependence of R_i (1) on T. Therefore one is able to examine the growth modes as a function of the misfit ε.

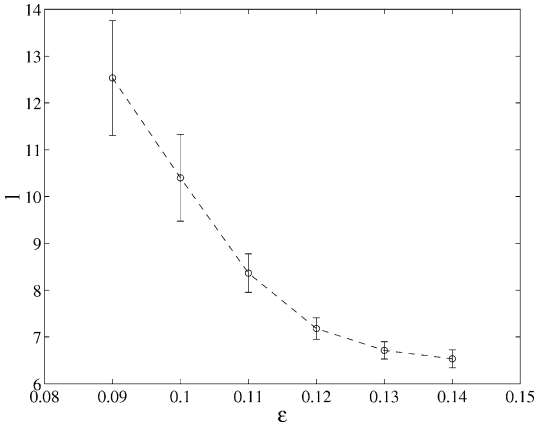

Fig. 3. Size of the monolayer island l at the 2D–3D transition as a function of ε. The error bars are obtained as the standard error of the simulation results.

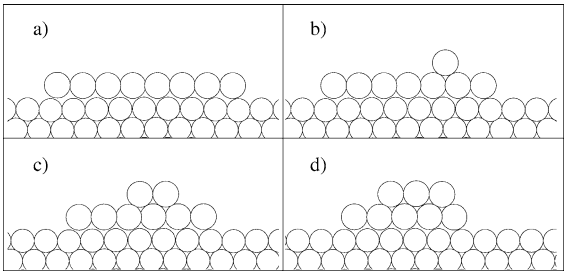

Fig. 4. 2D–3D transition observed in a simulation run with $\varepsilon = 0.11$. (a) Monolayer island, (b) particle from an edge manages to jump on the island, (c) another particle from the edge jumps on the island and forms a quite stable dimer with the first one, (d) a third particle from the edge completes the transition.

For simulations at temperature $T = 0.07U_0/k_B$ we find *layer-by-layer* growth for $-0.15 \leqslant \varepsilon \leqslant 0.08$ and 2D–3D transitions for $0.09 \leqslant \varepsilon \leqslant 0.14$. These transitions take place at a distinct island size l (number of particles in a monolayer island), depending on ε (Fig. 3): the smaller the misfit the bigger are the observed island sizes at the transition. Fig. 4 shows such a transition as it was observed in a simulation for $\varepsilon = 0.11$.

Transitions are also observed in simulations with deposition rate $R_d = 0$ and a fixed number of adsorbate particles. The 2D–3D transition is therefore not activated by the deposition of new particles on the monolayer island. But because of the misfit at distinct island sizes particles at the island edges are more weakly bound and the diffusion rates for a jump on the island are therefore increased [12].

5. Conclusion

We demonstrate that the proposed algorithm is applicable in the simulation of two different phenomena observed in heteroepitaxial growth: the appearance of misfit dislocations and the self-assembled island formation. Results on critical layer thickness and the island size at the 2D–3D transition are given. The algorithm will be applied to examine heteroepitaxial growth on vicinal surfaces and in $2 + 1$ dimensions.

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References

[1] P. Politi, G. Grenet, A. Marty, A. Ponchet, J. Villain, Phys. Rep. 324 (2000) 271–404.
[2] L. Dong, J. Schnitker, R.W. Smith, D.J. Sroloviz, J. Appl. Phys. 83 (1997) 217–227.
[3] D.A. Faux, G. Gaynor, C.L. Carson, C.K. Hall, J. Bernholc, Phys. Rev. B 42 (1990) 2914–2922.
[4] H. Spjut, D.A. Faux, Surf. Sci. 306 (1994) 233–239.
[5] W.M. Plotz, K. Hingerl, H. Sitter, Phys. Rev. B 45 (1992) 12 122–12 125.
[6] A. Schindler, Theoretical aspects of growth on one and two dimensional strained crystal surfaces, Dissertation, Duisburg, 1999.
[7] F. Much, M. Ahr, M. Biehl, W. Kinzel, Europhys. Lett. 56 (2001) 791–796.
[8] A. Barabasi, Appl. Phys. Lett. 70 (1997) 2565–2567.
[9] M.E.J. Newman, G.T. Barkema, Monte Carlo Methods in Statistical Physics, Oxford University Press, 1999.
[10] W.H. Press, S.A. Teukolsky, W.T. Vetterling, B.P. Flannery, Numerical Recipes in C, Cambridge University Press, 1992.
[11] G. Cohen-Solal, F. Bailly, M. Barbé, J. Crystal Growth 138 (1994) 68–74.
[12] F. Much, M. Ahr, M. Biehl, W. Kinzel, in preparation.